Division of Materials Chemistry
- Inorganic Photonics Materials -

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Prof HIMANSHU, Jain	Lehigh University, USA, 25–26 July 2006
Prof MCNAMARA, Pam	University of Sydney, Australia, 16 October 2006

Scope of Research

In this laboratory, amorphous and polycrystalline inorganic materials and organic-inorganic hybrid low-melting glassy materials with various optical functions such as photorefractivity, optical nonlinearity and photocatalysis are the target materials, which are synthesized by sol-gel, multi-cathode sputtering, melt-quenching and sintering methods and so on. Aiming at highly functional materials, the structures are investigated by X-ray diffraction techniques, high-resolution NMR, thermal analysis, various laser spectroscopies and ab initio molecular orbital calculations, and the properties are fully characterized.

Research Activities (Year 2006)

Presentations

Preparation and Application of Organic-inorganic Hybrid Low-melting Glass, Yoko T, Kyushu Synchrotron Photon Factory Completion Memory Symposium, Saga, 25 January 2006 (Invited).

Preparation and Properties of Novel Organic-inorganic Hybrid Low-melting Glasses, Mizuno M, Kakiuchida H, Tokuda Y, Takahashi M, Yoko T, HNM 2006 in Nagaoka, 4–5 February 2006 (Invited).

Preparation and Properties of Novel Organic-inorganic Hybrid Low-melting Glasses by a Non-aqueous Acid-base Reaction Method, Mizuno M, Tokuda Y, Takahashi M, Yoko T, International Symposium on Non Oxide and New Optical Glasses (ISNOG 2006), 10–14 April 2006, Bangalore, India (Invited).

Formation of Self-organized Ordered Micro Structures in Oxide Films through Photo-polymerization Induced Phase Separation, Takahashi M, Maeda T, Tokuda Y, Yoko T, IUMRS-ICA, Cheju, Korea, 16 September 2006.

Organic-inorganic Hybrid Materials through Solventless Processes, Takahashi M, Yoko T, The Ceramics Society of Japan Fall Meeting, Yamanashi, Japan, 19 September 2006 (Invited).

Organically-modified Silicophosphate Materials Prepared by Acid-base Reaction, Takahashi M, Yoko T, The Ceramics Society of Japan Fall Meeting, Yamanashi, Japan, 21 September 2006 (Invited).

Organically-modified Silicophosphate Hybrid Materials Prepared through Acid-base Concept, Takahashi M, Mizuno M, Tokuda Y, Yoko T, The XI International Conference on the Physics of Non-crystalline Solids, Rhodes, Greek, 2 November 2006.

Grants

Takahashi M, Development of Photonics Materials Based on the Organic-inorganic Hybrid Low Melting Glasses, PRESTO, Japan Science and Technology Agency, 1 November 2002–31 March 2006.

New Families of Organic-inorganic Hybrid Materials through Solventless Processes

Development of highly functional and reliable materials for optical information processing devices is one of the most important research targets these days. We have been reporting new families of organic-inorganic hybrid materials for such application prepared through solventless condensation processes, by which a flexible structural control at a molecular level is possible. For such purpose, we are working on the solventless condensation between the starting reagents through acid-base reaction and alcohol condensation. The obtained materials are characterized by a unique oxide alternating copolymer structure, enabling us to control the molecular structure for the molecular engineering precisely. Highly transparent and, if necessarily, patternable materials were obtained by these methods, and most of them were found to be suitable for photonics applications. Organically-modified silicophosphate glasses obtained through the acid-base reaction are found to be much better solvent for metal ions and ionic organic chromospheres than good organic solvent for them. It is expected for this materials that the figure-of-merit is improved and the device dimension could be largely reduced. The Au-nano particle- and rhodamine 6G-codoped phenyl-modified silicophasphate materials with 100% condensation yield, exhibited a large coefficient of two photon absorption which was 100 times as large as that of Au-nano particle-undoped one. In a case of alcohol condensation in solventless process, we have reported the organically-modified curable siloxane with extremely low optical attenuation, < 0.3 dB/cm, in the telecom window. Figure 1 is a sub-micrometer scale photonic structure obtained by the soft lithography using the present material.

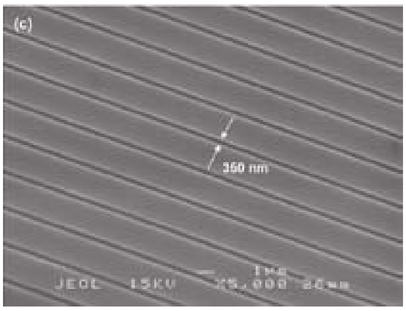

Figure 1. SEM image of photonic structure made of organically-modified siloxane materials obtained by solventless alcohol condensation of vinyltriisopropenoxysilane and diphenylsilanediol.

Inverse Methods for the MQMAS NMR Spectral Analysis

A number of structural studies on inorganic glass using experimental and simulation methods have been carried out. This type of study is important because the knowledge of the glass structure leads to an understanding of its physical and chemical properties. MAS NMR is one of the powerful tools to provide the structure information especially on dipolar nuclei (I = 1/2). Unfortunately, quadrupolar nuclei (I ≥ 3/2) provide too broad MAS NMR spectra to be analyzed quantitatively, although half elements have quadrupolar spin. For a better understanding of quadrupolar nuclei in solid state material, Fridman et al. developed MQMAS NMR to provide narrow spectra of quadrupolar nuclei. However, MQMAS NMR lacks quantitative information on the nuclei because the efficiency of multiquantum spin transition depends on the quadrupolar coupling constant.

In this work, we will provide a direct investigation of the local structure in inorganic glass based both on MQMAS NMR and inverse analysis. An observed spectrum and an inherent distribution of structure parameter, R, are related with each other by

$$I(\varpi) = \int I_0(\varpi;R)\Pi(R)dR \qquad (1)$$

where $I(\varpi)$, $I_0(\varpi;R)$, $\Pi(R)$ are the observed spectrum, a theoretical spectrum for R, the distribution of R, respectively. Using equation (1), $\Pi(R)$ can be calculated based on the numerical approach with Tikhonov regularization (inverse analysis). We are now trying to establish inverse analysis to extract the inherent structure distribution in inorganic glass from the measured NMR spectra.

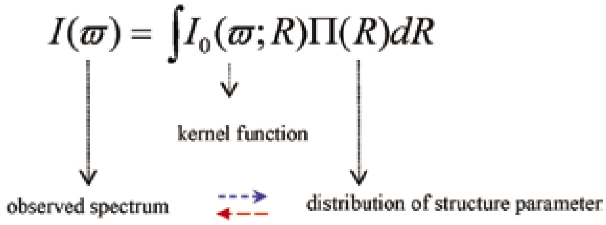

Figure 2. Inverse method for quantitative analysis on MQMAS NMR spectra. The arrows, ← and →, mean direct and inverse problem, respectively. The kernelfunction can be calculated theoretically using quantum theory.

Takahashi M, Inhomogeneous Structures in the Glasses, Grant-in-Aid for Scientific Research for Encouragement of Young Scientists (A), 1 April 2004–30 September 2006.

Takahashi M, Fabrication of Large Area Photonic Films, Toyota Physical & Chemical Research Institute, 1 April 2005–31 March 2006.

Tokuda Y, Fabrication of Pb-free Sealing Glass, JST, Research for Promoting Technological Seeds, 13 September 2006–28 February 2007.